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PATENT SPECIFICATION

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(21) Application No. 51961/77 (31) Convention Application No. 2656747

(22) Filed 14 Dec. 1977

(32) Filed 15 Dec. 1976 in

(33) Federal Republic of Germany (DE)

(44) Complete Specification published 17 June 1981

(51) INT CL3 C07D 265/30 A01N 9/22 A61K 31/535 C07D 295/02

(52) Index at acceptance

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C2C 1562 215 246 247 255 25Y 29X 29Y 776 802 805 80Y AA



(71) We, BASF AKTIENGESELLSCHAFT, a German Joint Stock Company of 6700 Ludwigshafen, Federal Republic of Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:-

The present invention relates to new and valuable morpholine derivatives and their salts and molecular compounds and adducts, having a good fungicidal action, fungicides containing these compounds, and processes for combatting fungi with these compounds.

The use of N - tridecyl - 2,6 - dimethylmorpholine and its salts, molecular compounds and adducts as fungicides has been disclosed (German Patent 1.164.152, German Patent 1,173,722, German Laid-Open Application DOS 2,461,513).

We have now found that morpholine derivatives of the formula

where R1, R2, R3 and R4 denote hydrogen, methyl or ethyl, and their salts,

molecular compounds and adducts have a good fungicidal action.

The results tabulated in Examples 3—5 below show that the fungicidal action of compounds according to the invention is superior to that of prior art commercial morpholine derivatives.

Examples of salts are those with inorganic or organic acids, e.g., chlorides, fluorides, bromides, iodides, sulfates, nitrates, phosphates, acetates, propionates and fumarates. Molecular compounds and adducts are formed for instance with acids of surfactants, e.g., dodecylbenzenesulfonic acid.

If the new compounds are 2,6-dimethyl derivatives, they may be isolated as cis and trans isomers. The compounds may be manufactured for instance by reaction of a morpholine of the formula

where R1 to R4 have the above meanings, e.g. 2,6 - dimethylmorpholine, with 3 p - tertbutylphenyl - 2 - methylpropanal having the formula

in the presence of a diluent, e.g. formic acid, at from 50° to 110°C. The preparation of the new compounds is illustrated by the following examples. 5 **EXAMPLE 1** Š Synthesis of N-(3-p-tert-butylphenyl-2-methyl-1-propyl)cis-2,6-dimethylmorpholine 2,6-Dimethylmorpholine which has been obtained by sulfuric acid-catalyzed cyclization of diisopropanolamine is separated into the cis and trans forms by fractional distillation in a column packed with steel mesh coils. About 75 wt% of 10 10 the 2,6-dimethylmorpholine is in the cis configuration, the remainder in the trans form. The isomers are separated by fractional distillation in a column having about 40 theoretical plates. The cis form distils as a 99% product at 80° to 81°C and a pressure of 100 mm Hg. The trans form of 2,6-dimethylmorpholine may be obtained under the same conditions at 88° to 89°C/100 mm Hg as a more than 95% 15 15 , product. 575 g of 98% formic acid is placed in a stirred apparatus equipped with reflux condenser, thermometer and dropping funnel. While stirring and cooling, 345 g of 99% 2,6-cis-dimethylmorpholine is then dripped in. The mixture is subsequently slowly heated on a water bath to 70°C. Over a period of 4 hours, 612 g of 3-p-tert-butylphenyl-2-methylpropanal is dripped into the reaction mixture, a temperature 20 201 of approx. 100°C being maintained. Condensation proceeds with marked evolution of CO₂. Upon conclusion of the reaction the mixture is kept for 2 hours at 100°C while stirring. The excess formic acid is then distilled off under reduced pressure. Substantial 25 25 separation of the formic acid is achieved under a water pump vacuum at 100°C. The base is liberated from the formate by dripping in 500 g of 40% aqueous caustic soda solution. The caustic soda solution is advantageously added at from 80° to 100°C to facilitate admixture of the amine phase which forms with the caustic phase. 200 g of toluene is added to depress the viscosity. After separation of -30 30 the caustic phase, the organic phase is washed twice with water, each time with 250 For further purification, the amine is fractionally distilled at 0.2 mm Hg in a distillation column having 5 trays. In addition to a small amount of first runnings (up to $143^{\circ}\text{C}/0.2$ mm Hg; 50 g), there is obtained 865 g of N - (3 - p - tert - butylphenyl) - 2 - methyl - 1 - propyl) - 2,6 - cis - dimethylmorpholine, which distils at 0.2 mm Hg between 143° and 146°C. According to gas-chromatographic analysis, the amine is more than 98% pure. With reference to aldehyde, the yield 35 35 40 To convert the compound to the hydrochloride, 30 g of the pure product is 40 dissolved in 50 g of ethanol which has been saturated with hydrogen chloride at room temperature. After cooling, there is obtained 23 g of the hydrochloride (m.p. 220°C) in very pure form. **EXAMPLE 2** 45 Synthesis of N-(3-p-tert-butylphenyl-2-methyl-1-propyl)-45 2,6-trans-dimethylmorpholine While cooling with ice, 29 g of 2,6-trans-dimethylmorpholine is introduced into 70 g of 98% formic acid, while stirring, 41 g of 3 - p - tert - butylphenyl - 2 - methylpropanal is then added. The reaction mixture is heated for 6 hours at 100°C. 50 Upon commencement of the reaction, very strong CO2 evolution takes place which 50 drops noticeably after about an hour. Further working up is effected as disclosed in more detail in Example 1. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,6 - trans dimethylmorpholine distils at 168°—169°C (5 mm Hg). The yield is 52 g (86% of theory, with reference to aldehyde). 11 g of the amine is dissolved in 20 g of ethyl 55 55 acetate which has been saturated with anhydrous hydrogen chloride. The hydrochloride crystallizes out upon cooling; m.p.: 165°C

The other compounds according to the invention may be obtained

analogously.

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Examples of compounds according to the invention are given below:

$$R = CH_3 - CH_2 - CH_$$

Compound no.

b.p.: 123°C/0.01 mm

trans form

				7
	12	R-N O CH ₃	b.p.: 125 ⁰ C/0.02 mm	
	13	R-N 3	b.p.: 170°C/5.0 mm	
	14	R-N HC1	m.p.: 206°C	
	15	R-N H ₃ C	b.p.: 145°C/O.3 mm	
5	16	H ₃ C R-N 0 . HC1	m.p.: 172°C	5
	17:	C ₂ H ₅ R-N O C ₂ H ₅	b.p.: 148°C/O.3 mm	
	18	R-N O C2H5	b.p.: 146-148°C/0.3 mm	
	19	C ₂ H ₅ CH ₃ R-N O .HBr	m.p.: 228°C	
10	20	cis form CH ₃ R-N O .HOOC - CH CH ₃ HC - COOH cis form	m.p.: 174°C	10
	•			

The morpholine derivatives according to the invention and the fungicides containing them are particularly suitable for combatting plant diseases such as Erisiphe graminis in cereals, Erysiphe cichoriacearum in Cucurbitaceae, Podosphaera leucotricha in apples, Uncinula necator in grapes, Erysiphe polygoni in beans, Sphaerotheca pannosa in roses, Microsphaera querci in oaks, Botrytis cinerea in strawberries and grapes, Mycosphaerella musicola in bananas, Puccinia species (rusts) in cereals, Uromyces appendiculatus and U. phaseoli in beans, Hemileia vastatrix in coffee, and Rhizoctonia solani. Therefore, they may be applied, for example, to cereal plants (e.g. wheat and barley for protection against rust and/or mildew), bean plants, apple trees, grape vines, cucurbitaceae plants, rose trees, oak trees, strawberry plants, banana trees and coffee plants. They have a systemic action; they are not only taken up by the roots but also absorbed by the leaves, and are translocated in the plant tissue. They may be used to treat the plants after fungus attack has begun.

Application is effected for instance by watering, spraying, dusting, or disinfecting the plants or treating the seed with the morpholine derivatives.

	When the morpholine derivatives are used to protect plants against fungus	
	infection, the application rates are from 0.025 to 5 kg of active ingredient per	
	hectare. For the surface protection of trees or fruit, the morpholine derivatives	
	may also be used in combination with plastics dispersions in amounts of from 0.25	
5.	to 5%, based on the weight of the dispersion. In general, the fungicidal	5
٦.	compositions according to the invention contain from 0.1 to 95, preferably from 0.5	
	to 90, wt% of the morpholine derivative.	
	The morpholine derivatives may also be mixed with other, prior art,	
	fungicides. In many instances, the spectrum of fungicidal action is increased; with a	
10	number of fungicidal compositions in the weight ratio range of 1:10 to 10:1	10
10	synergistic effects also occur, i.e., the fungicidal action of the combination product	
	is greater than the effect of the individual components added together. Examples of	
	fungicides which may be combined with the morpholine derivatives of the	
	invention are dithiocarbamates and derivatives thereof, e.g.,	
	milenting give control of the contro	
15	zinc dimethyldithiocarbamate	15
	manganese ethylenebisdithiocarbamate	
	zinc ethylenebisdithiocarbamate	
	tetramethylthiuram disulfide	
	ammonia complex of zinc-(N,N'-ethylene-bisdithiocarbamate) and	
20	N.N'-polvethylene-bis-(thiocarbamovl)-disulfide	20
, ,	ammonia complex of zinc-(N,N'-propylene-bisdithiocarbamate) and	
	N,N'-polypropylene-bis-(thiocarbamoyl)-disulfide	
:		•
	heterocyclic structures, such as	
•	N-trichloromethylthiotetrahydrophthalimide	2.5
25	N-trichloromethylthiophthalimide	25
	N-(1,1,2,2-tetrachloroethylthio)-tetrahydrophthalimide	
	methyl 1-(butylcarbamoyl)-2-benzimidazole carbamate	
• •	2-methoxycarbonylaminobenzimidazole	
- 20	2,3-dihydro-5-carboxanilido-6-methyl-1,4-oxathiin-4,4-dioxide 2,3-dihydro-5-carboxanilido-6-methyl-1,4-oxathiin	30
30	5-butyl-2-dimethylamino-4-hydroxy-6-methylpyrimidine	30
	1,2-bis-(3-ethoxycarbonyl-2-thioureido)-benzene	
٠.	1,2-bis-(3-methoxycarbonyl)-2-thioureido)-benzene	•
•	1,2-bis-(3-methoxyeuroony), 2 chiedroids, comment	
	and various fungicides, such as	
: 35	dodecylguanidine acetate	35
	N-dichlorofluoromethylthio-N',N'-dimethyl-N-phenylsulfuric acid diamide	
	2,5-dimethylfuran-3-carboxylic acid anilide	
	2,5-dimethylfuran-3-carboxylic acid cyclohexyl amide	
	2-iodobenzoic acid anilide	. 40
40	2-bromobenzoic acid anilide	. 40
	3-nitroisophthalic acid diisopropyl ester	•
	1-(1,2,4-triazolyl-1')-[1-(4'-chlorophenoxy)]-3,3-dimethylbutan-2-one	•
	1-(1-imidazolyl)-2-allyloxy-2-(2,4-dichlorophenyl)-ethane	
	piperazine-1,4-diyl-bis-1-(2,2,2-trichloroethyl)-formamide 2,4,5,6-tetrachloroisophthalonitrile	45
45	1,2-dimethyl-3,5-diphenylpyrazoliniummethylsulfate.	45
	1,2-difficting1-3,3-dipficinglypy1azonimummethy.sunate.	
	Application may be effected for instance in the form of directly sprayable	
	solutions, powders, suspensions, dispersions, emulsions, oil dispersions, pastes,	
	dusts, broadcasting agents, or granules by spraying, atomizing, dusting,	
50	broadcasting or watering. The forms of application depend entirely on the purpose	50
50	for which the morpholine derivatives are being used; in any case they should ensure	
	a fine distribution of the morpholine derivative.	
	For the preparation of solutions, emulsions, pastes and oil dispersions to be	
•	sprayed direct, mineral oil fractions of medium to high boiling point, such as	
55	kerosene or diesel oil, further coal-tar oils, and oils of vegetable or animal origin,	55
	aliphatic, cyclic and aromatic hydrocarbons such as benzene, toluene, xylene,	
	paraffin tetrahydronaphthalene and alkylated naphthalenes, and hydrocarbon	
	derivatives such as methanol, ethanol, propanol, butanol, chloroform, carbon	
	tetrachloride, cyclohexanol, cyclohexanone, chlorobenzene and isophorone, as	

well as strongly polar solvents such as dimethylformamide, dimethyl sulfoxide, Nmethylpyrrolidone and water, are suitable. Aqueous formulations may be prepared from emulsion concentrates, pastes, oil dispersions or wettable powders by adding water. To prepare emulsions, pastes 5 and oil dispersions the ingredients as such or dissolved in an oil or solvent may be 5 homogenized in water by means of wetting or dispersing agents, adherents or emulsifiers. Concentrates which are suitable for dilution with water may be prepared from the morpholine derivative wetting agent, adherent, emulsifying or dispersing agent and possibly solvent or oil. 10 Examples of surfactants are: alkali metal, alkaline earth metal and ammonium 10 salts of ligninsulfonic acid, naphthalenesulfonic acids, phenolsulfonic acids, alkylaryl sulfonates, alkyl sulfates, and alkyl sulfonates, alkali metal and alkaline earth metal salts of dibutylnaphthalenesulfonic acid, lauryl ether sulfate, fatty alcohol sulfates, alkali metal and alkaline earth metal salts of fatty acids, salts of 15 sulfated hexadecanols, heptadecanols, and octadecanols, salts of sulfated fatty 15 alcohol glycol ethers, condensation products of sulfonated naphthalene and naphthalene derivatives with formaldehyde, condensation products of naphthalene or naphthalenesulfonic acids with phenol and formaldehyde, polyoxyethylene octylphenol ethers, ethoxylated isooctylphenol, ethoxylated octylphenol and ethoxylated nonylphenol, alkylphenol polyglycol ethers, tributylphenol polyglycol 20 20 ethers, alkylaryl polyester alcohols, isotridecyl alcohols, fatty alcohol ethylene oxide condensates, ethoxylated castor oil, polyoxyethylene alkyl ethers, ethoxylated polyoxypropylene, lauryl alcohol polyglycol ether acetal, sorbitol esters, lignin, sulfite waste liquors and methyl cellulose. Powders, dusts and broadcasting agents may be prepared by mixing or grinding the morpholine derivatives with a solid carrier. 25 25 Granules, e.g., coated, impregnated or homogeneous granules, may be prepared by bonding the morpholine derivatives to solid carriers. Examples of solid carriers are mineral earths such as silicic acid, silica gels, silicates, talc, kaolin, 30 Attaclay, limestone, lime, chalk, bole, loess, clay, dolomite, diatomaceous earth, 30 calcium sulfate, magnesium sulfate, magnesium oxide, ground plastics, fertilizers such as ammonium sulfate, ammonium phosphate, ammonium nitrate, and ureas, and vegetable products such as grain flours, bark meal, wood meal, and nutshell meal, cellulosic powders, etc. There may be added to the compositions or individual active ingredients oils of 35 35 various types, herbicides, fungicides, nematocides, insecticides, bactericides, trace elements, fertilizers, antifoams (e.g., silicones), growth regulators, antidotes and other herbicidally effective compounds. The following experiments, the following prior art compounds were used for 40 comparison purposes: 40 Compound no. 7. (prior art) 8 45 45 (prior art) 9

(prior art)

The BOOK Statement of the

EXAMPLE 3

	EXAMPLE 3 Leaves of wheat seedlings of the "Jubilar" variety grown in pots are treated with aqueous emulsions consisting of 80% (wt%) of active ingredient and 20% emulsifier and dusted, after the sprayed-on layer has dried, with spores of wheat	
5	mildew (Erysiphe graminis var. tritici). The plants are then placed in a greenhouse at from 20° to 22°C and 75 to 80% relative humidity. The extent the fungus has spread is determined after 10 days.	5
	Leaf Attack After Spraying With Liquors	
10	Containing Active Ingredients in Amounts of Active Ingredient 0.006% 0.012% 0.025% 0.05%	10
	1 1—2 1 0—1 0	
	$\begin{array}{cccccccccccccccccccccccccccccccccccc$	
	4 i 0 0 0	
15	$egin{array}{cccccccccccccccccccccccccccccccccccc$	15
	19 0 0 0	
	$\begin{bmatrix} 20 & 0 & 0 & 0 & 0 \\ 7 & 3 & 4 & 3 & 2 & 1 \end{bmatrix}$	
20	8 > prior art 4 4 2 1	20
•	9	•
	0=no attack, graduated down to 5=surface of leaves completely covered by fungus.	
25	EXAMPLE 4	25
	Leaves of barley seedlings of the "Firlbecks Union" variety grown in pots are treated as described in Example 3, and dusted with spores of barley mildew (Erysiphe graminis var. hordei).	
20	Leaf Attack After Spraying With Liquors	30
30	Containing Active Ingredient in Amounts of	30
30	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0	30
30	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	30
30	Containing Active Ingredient in Amounts of Active Ingredient 0.006% 0.012% 0.025% 1 0 0 0 2 0 0 0 3 0 0 0 4 0 0	30 35
30	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 2 0 0 0 3 0 0 0 4 0 0 0 5 0 0 0 6 0 0 0	
30	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 2 0 0 0 3 0 0 0 4 0 0 0 5 0 0 0 6 0 0 0	
30 35	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	
	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 2 0 0 0 3 0 0 0 4 0 0 5 0 0 0 0 6 0 0 0 19 0 0 0 19 0 0 0 20 0 0 0 7 } prior art 3 1 1 1	35
	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1	35
	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 2 0 0 0 3 0 0 0 4 0 0 5 0 0 0 0 6 0 0 0 19 0 0 0 19 0 0 0 20 0 0 0 7 } prior art 3 1 1 1	35
	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	35
40	Containing Active Ingredient in Amounts of 0.006% 0.012% 0.025% 1 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0 0	35 40

8		1,591,	267		8
	Leaf Attack After Spraying With Liquor Containing Active Ingredient in Amounts of 0.025 0.05 0.1				
5	1 3	2—3	1—2 0	. 0.1 0 0	5
	4 5 6	0 0	0 0	0 0	,
10	19 20	0 0 0	0 0 0	0 0 0	10
	7 8 9 prior art	4 4 4	0 3 4 · 3	2 3 3	
	Control (untreated)	•	4	11,	· · · · ·
15	0=no damage, graduated of fungus.	lown to 5=sur	face of leaves com	pletely covered by	15
20	90 parts by weight of commethyl- α -pyrrolidone. A mixtur form of very fine drops.	EXAMPL npound 3 is re is obtained	mixed with 10 part	s by weight of N- application in the	
	torm of vory line grops.	EXAMPL	E 7	•	, 20
25	20 parts by weight of compound 4 is dissolved in a mixture consisting of 80 parts by weight of xylene, 10 parts by weight of the adduct of 8 to 10 moles of ethylene oxide to 1 mole of oleic acid N-monoethanolamide, 5 parts by weight of the calcium salt of dodecylbenzenesulfonic acid, and 5 parts by weight of the adduct of 40 moles of ethylene oxide to 1 mole of castor oil. By pouring the solution into 100,000 parts by weight of water and uniformly distributing it therein, an aqueous dispersion is obtained containing 0.02% by weight of the active ingredient.				
30 35	EXAMPLE 8 20 parts by weight of compound 3 is dissolved in a mixture consisting of 40 parts by weight of cyclohexanone, 30 parts by weight of isobutanol, 20 parts by weight of the adduct of 7 moles of ethylene oxide to 1 mole of isooctylphenol, and 10 parts by weight of the adduct of 40 moles of ethylene oxide to 1 mole of castor oil. By pouring the solution into 100,000 parts by weight of water and uniformly distributing it therein, an aqueous dispersion is obtained containing 0.02% by weight of the active ingredient.				
40	20 parts by weight of com parts by weight of cyclohexanol a boiling point between 210° an moles of ethylene oxide to 1 mol parts by weight of water and dispersion is obtained containing	EXAMPLI pound 4 is di , 65 parts by v d 280°C, and le of castor oil	ssolved in a mixtur veight of a mineral of 10 parts by weight of By pouring the sold distributing it the	oil fraction having of the adduct of 40 ution into 100,000	40
45	20 parts by weight of comp sodium salt of diisobutylnaphtl sodium salt of a ligninsulfonic parts by weight of powdered uniformly distributing the mixtuis obtained containing 0.1% by	iaiene-α-suifo acid obtained silica gel, an re in 20 000 ps	I mixed with 3 parts nic acid, 17 parts from a sulfite was d triturated in a larts by weight of we	by weight of the te liquor, and 60	45 50
55	3 parts by weight of compour particulate kaolin. A dust is oingredient.	EXAMPLE and 3 is intima btained cont	11 tely mixed with 97 paining 3% by weig	parts by weight of the active	50
	30 parts by weight of compo of 92 parts by weight of powder which has been sprayed onto the active ingredient is obtained ha	red silica gel he surface of	ately mixed with a n and 8 parts by weig this silica gel. A fo	ht of paraffin oil	55

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EXAMPLE 13

40 parts by weight of compound 3 is intimately mixed with 10 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate, 2 parts of silica gel and 48 parts of water to give a stable aqueous dispersion. Dilution in 100,000 parts by weight of water gives an aqueous dispersion containing 0.04 wt% of active ingredient.

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EXAMPLE 14

20 parts of compound 4 is intimately mixed with 2 parts of the calcium salt of dodecylbenzenesulfonic acid, 8 parts of a fatty alcohol polyglycol ether, 2 parts of the sodium salt of a phenolsulfonic acid-urea-formaldehyde condensate and 68 parts of a paraffinic mineral oil. A stable oily dispersion is obtained.

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WHAT WE CLAIM IS:-

1. A morpholine derivative of the formula

where R1, R2, R3 and R4 each independently denote hydrogen, methyl or ethyl, or a 15 15 salt, molecular compound or adduct thereof. 2. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2.6dimethylmorpholine. 3. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2 -20 methylmorpholine. 20 4. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3methylmorpholine. 5. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - morpholine. 6. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,2,6 -25 trimethylmorpholine. 25 7. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2.5 - 1 - 2.5 - 2.dimethylmorpholine. 8. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3,5 dimethylmorpholine. 9. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 2,6 -. 30 30 diethylmorpholine. 10. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - 3.5 - 10.diethylmorpholine. 11. N - (3 - p - tert - butylphenyl - 2 - methyl - 1 - propyl) - cis - 2,6 -35 35 dimethylmorpholine. 12. A morpholine derivative as claimed in any of claims 1 to 11 in the form of a molecular compound or adduct with an acid of a surfactant or in the form of a salt with an inorganic or organic acid. 13. A morpholine derivative as claimed in claim 12 in the form of its chloride, fluoride, bromide, iodide, sulfate, nitrate, phosphate, acetate, propionate or 40 40 fumarate salt or its dodecylbenzenesulfonic acid adduct or molecular compound. 14. A process for preparing a morpholine derivative as claimed in claim 1 wherein 3 - p - tert - butylphenyl - 2 - methylpropanal of the formula



where R1, R2, R3 and R4 have the meanings given in claim 1, in the presence of a diluent at a temperature of from 50° to 110°C. 15. A morpholine derivative as claimed in claim I when prepared by a process 5 as claimed in claim 14, or a salt, molecular compound or adduct thereof. 5 16. The use of a compound as claimed in any of claims 1 to 13 or 15 as a fungicide (other than for application to human beings). 17. A fungicidal composition comprising a solid or liquid carrier and a compound as claimed in any of claims 1 to 13 or 15. 18. A fungicidal composition as claimed in claim 17 in the form of a solution, 10 10 emulsion, paste or oil dispersion in an organic liquid to be sprayed direct or diluted with water for spraying. 19. A fungicidal composition as claimed in claim 17 as an aqueous formulation. 20. A fungicidal composition comprising a compound as claimed in any of claims 1 to 13 or 15 and a wetting or dispersing agent, adherent or emulsifier. 15 15 21. A fungicidal composition as claimed in claim 17 in the form of a dust, wettable powder, broadcastable composition or granules. 22. A fungicidal composition as claimed in any of claims 17 to 21 which contains a further fungicide. 20 23. A fungicide composition comprising a compound as claimed in any of 20 claims 1 to 13 or 15 and a further fungicidal compound listed herein. 24. A process for preparing a fungicidal composition, wherein a solid or liquid carrier is mixed with a compound as claimed in any of claims 1 to 13 or 15. 25. A process for combatting fungi, wherein the locus to be protected against 25 fungus attack (which locus is not a human being) is treated with a compound as 25 claimed in any of claims 1 to 13 or 15. 26. A process for combatting fungal attack in plants wherein the plants, or their seeds, are treated with a compound as claimed in any of claims 1 to 13 or 15. 27. A process as claimed in claim 26 wherein the plants are treated after fungus 30 attack. 30 28. A process as claimed in claim 26 or 27 wherein the plants are cereal plants, bean plants, apple trees, grape vines, cucurbitaceae plants, rose trees, oak trees, strawberry plants, banana trees, or coffee plants. 29. A process as claimed in claim 28 wherein wheat or barley plants are treated 35 for protection against rust and/or mildew. 35

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Printed for Her Majesty's Stationery Office, by the Courier Press, Learnington Spa, 1981 Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.